

Spin-Ordered States in Multilayer Massless Dirac Fermion Systems

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We investigate the spin-ordered states in multilayer massless Dirac fermion systems under magnetic fields, in which the intralayer interaction is ferromagnetic owing to the exchange interaction, while the interlayer interaction is antiferromagnetic arising from the interlayer hopping and the on-site Coulomb repulsion. The possible spin-ordered states are examined within the mean field theory, and we apply it to α -(BEDT-TTF)₂I₃, which is a multilayer massless Dirac fermion system under pressure. In the weak interlayer coupling regime the system exhibits a ferromagnetically spin-ordered state with the effective Zeeman g -factor less than two contrasting to that observed in the single-layer graphene.

A multilayer organic conductor α -(BEDT-TTF)₂I₃ [BEDT-TTF=bis(ethylenedithio)tetrathiafulvalene]¹ has attracted a great deal of attraction since it was found that the energy dispersion is linear under pressure.²⁻⁴ α -(BEDT-TTF)₂I₃ has a layered structure, in which conducting layers of BEDT-TTF molecules and insulating layers of I₃ anions stack alternatively. Below 135 K, charge ordering with a stripe pattern makes α -(BEDT-TTF)₂I₃ an insulating phase under ambient pressure.⁵⁻⁸ For pressures higher than 1.5GPa, the charge ordering transition is suppressed and the system becomes metallic even at low temperature.⁹ The resistivity is almost temperature independent while the Hall coefficient shows strong temperature dependence.⁹

Using the tight-binding model with the transfer integrals obtained by an X-ray diffraction experiment,¹⁰ Kobayashi and coworkers calculated the energy dispersion of α -(BEDT-TTF)₂I₃.^{2,3} They found that the band structure near the Fermi energy is described by a tilted and anisotropic Dirac cone, which was supported by the first principles calculation.^{11,12} The presence of Dirac fermions is clearly demonstrated in the interlayer magnetoresistance measurement¹³ where the zero energy Landau level of Dirac fermions leads to negative magnetoresistance.¹⁴

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In graphene,¹⁵ which is a well-established Dirac fermion system, the existence of the Dirac fermion spectrum was clearly demonstrated by the observation of the half-integer quantum Hall effect.^{16,17} Under a high magnetic field, lifting of spin degeneracy is observed experimentally.¹⁸ Nomura and MacDonald examined a criterion for the occurrence of quantum Hall ferromagnet states at zero-temperature.¹⁹ Under a magnetic field, the kinetic energy is quenched into the Landau levels while the Landau level broadening plays the role of the band width. In order to stabilize a quantum Hall ferromagnetic state, a cleaner system is plausible. In this regard, we expect that a quantum Hall ferromagnetic state is more stable in α -(BEDT-TTF)₂I₃ than in graphene since the former is cleaner than the latter.¹³ We also expect that the multilayer structure of α -(BEDT-TTF)₂I₃ should lead to symmetry broken states at finite temperature.

In this work, we investigate the possible spin-ordered state in α -(BEDT-TTF)₂I₃ within the mean field theory. In our model, the intralayer ferromagnetic interaction arises from the exchange interaction and the interlayer antiferromagnetic interaction arises from interlayer hopping and on-site Coulomb repulsion. We also include the Zeeman energy term that plays an important role in selecting a stable spin-ordered state.

We study multilayer massless Dirac fermion system under a magnetic field. In each layer, we consider a single component of Dirac fermions. In general there are two Dirac points in the Brillouin zone. Here, we assume that Dirac fermions are degenerate with respect to these valley degrees of freedom and we do not consider the possibility of lifting valley degeneracy. For the description of Dirac fermions in each layer, we take the following Hamiltonian:

$$H = v \begin{pmatrix} 0 & (p_x + eA_x) - i(p_y + eA_y) \\ (p_x + eA_x) + i(p_y + eA_y) & 0 \end{pmatrix},$$

where p_α and A_α with $\alpha = x, y$ are momentum operators and the vector potential, respectively. The velocity of Dirac fermions is denoted as v and $-e$ is the electron charge. We take the Landau gauge, $A_x = 0$ and $A_y = Bx$, with B being the applied magnetic field. In α -(BEDT-TTF)₂I₃, the energy dispersion of Dirac fermions is described by a tilted and anisotropic cone.³ However, under a magnetic field, tilting and anisotropy introduce a renormalization of the velocity v .^{20,21} Thus, we assume that this renormalization effect is already included in v . For the case of α -(BEDT-TTF)₂I₃, we take $v = 10^7$ cm/s.¹³

Taking the plane wave form with the wave number k in the y -direction, the Landau level wave functions for Dirac fermions are given by

$$\psi_{n,k}(x, y) = \frac{1}{\sqrt{L}} \exp(iky) \phi_{n,k}(x), \quad (1)$$

with L being the system dimension. The energy spectrum is $E_n = \text{sgn}(n) \sqrt{2|n|} \hbar v / \ell_B$. Here n , the Landau level index, is an integer and $\ell_B = \sqrt{\hbar / (eB)}$ is the magnetic length. The function $\phi_{n,k}(x)$ is given by

$$\phi_{n,k}(x) = \frac{C_n}{\sqrt{\ell_B}} \left[\begin{pmatrix} -i \text{sgn}(n) \\ 0 \end{pmatrix} h_{|n|-1} \left(\frac{x}{\ell_B} + k \ell_B \right) + \begin{pmatrix} 0 \\ 1 \end{pmatrix} h_{|n|} \left(\frac{x}{\ell_B} + k \ell_B \right) \right]. \quad (2)$$

Here, $h_{|n|}(\xi)$ is the harmonic oscillator wave function and the normalization constant C_n is $C_0 = 1$ and $C_n = 1 / \sqrt{2}$ for $n \neq 0$. In terms of these Landau level wave functions, the electron field operator is written as

$$\hat{\psi}(x, y) = \sum_{n,k,\sigma} \psi_{n,k}(x, y) \hat{c}_{n,k,\sigma}, \quad (3)$$

where $\hat{c}_{n,k,\sigma}$ is the annihilation operator of Dirac fermions with the Landau level index n , the wave number k , and spin σ . The density operator is defined by $\hat{\rho}(\mathbf{r}) = \hat{\psi}^\dagger(x, y) \hat{\psi}(x, y)$. The Fourier transform of $\hat{\rho}(\mathbf{r})$ is

$$\hat{\rho}_{\mathbf{q}} = \int d^2\mathbf{r} \exp(-i\mathbf{q} \cdot \mathbf{r}) \hat{\rho}(\mathbf{r}) = \sum_{n,n',k,\sigma} F_{n,n',k}^{\mathbf{q}} c_{n,k,\sigma}^\dagger c_{n',k+q_y,\sigma}, \quad (4)$$

where $F_{n,n',k}^{\mathbf{q}}$ is the Landau level form factor. Using the density operator, the Coulomb interaction is described as

$$V_C = \frac{1}{2L^2} \sum_{\mathbf{q}} V_{\mathbf{q}} \hat{\rho}_{\mathbf{q}} \hat{\rho}_{-\mathbf{q}}, \quad (5)$$

where $V_{\mathbf{q}} = e^2 / (2\epsilon q)$ with ϵ being the dielectric constant.

Now we introduce the mean field approximation for the exchange interaction:²²

$$\begin{aligned} V_C^{MF} = & -\frac{1}{2L^2} \sum_{\mathbf{q}} V_{\mathbf{q}} \left(\sum F_{n_2,n_2,k}^{\mathbf{q}} F_{n_1,n_2,k+q_y}^{-\mathbf{q}} \langle c_{n_2,k+q_y,\sigma}^\dagger c_{n_2,k+q_y,\sigma} \rangle c_{n_1,k,\sigma}^\dagger c_{n_2,k,\sigma} \right. \\ & \left. + \sum F_{n_1,n_2,k}^{\mathbf{q}} F_{n_1',n_1,k+q_y}^{-\mathbf{q}} \langle c_{n_1,k,\sigma}^\dagger c_{n_1,k,\sigma} \rangle c_{n_1',k+q_y,\sigma}^\dagger c_{n_2,k+q_y,\sigma} \right). \end{aligned} \quad (6)$$

In the following, we consider α -(BEDT-TTF)₂I₃, and we assume that the Fermi energy is at the Dirac point. In this case, the zero energy Landau level, the presence of which is a characteristic feature of Dirac fermions, is at the Fermi energy. We may consider only the zero energy Landau level since the $n = 1$ Landau level energy, $E_1 \simeq 10 \sqrt{B}$ is large enough compared with the Landau level width at low temperatures.²³ The Landau level mixing is important at high temperatures. For instance, the Landau level mixing is not negligible for $T > 10$ K at $B = 1$ T. However, we are interested in low temperature behaviors and we do not consider the Landau level mixing.

The mean field Hamiltonian for the zero energy Landau level is

$$H_0^{MF} = \sum_{k,\sigma} \left[\varepsilon_k - \frac{1}{L^2} \sum_{\mathbf{q}} V_{\mathbf{q}} \exp\left(-\frac{q^2 \ell_B^2}{2}\right) \langle c_{k+q_y,\sigma}^\dagger c_{k+q_y,\sigma} \rangle \right] c_{k,\sigma}^\dagger c_{k,\sigma}, \quad (7)$$

where we have used $F_{0,0,k}^{\mathbf{q}} F_{0,0,k+q_y}^{-\mathbf{q}} = \exp(-q^2 \ell_B^2/2)$. In eq. (7) we introduce k -dependent energy ε_k in order to introduce the broadening of the Landau level in the presence of disorder. For simplicity, we assume that the density of states of the Landau level has the following form suggested from the self-consistent Born approximation (SCBA):^{22,24}

$$D(\varepsilon) = \frac{4}{\pi\Gamma} \sqrt{1 - \left(\frac{2\varepsilon}{\Gamma}\right)^2}, \quad (8)$$

with Γ being the Landau level width. According to the SCBA, under high magnetic fields, Γ is proportional to \sqrt{B} .^{22,24} However, for the reason we shall explain below, we regard Γ as a constant. Within the mean field approximation, the electron self-energy satisfies the following self-consistent equation:

$$\Sigma_\sigma = -\frac{1}{L^2} \sum_{\mathbf{q}} V_{\mathbf{q}} \exp\left(-\frac{q^2 \ell_B^2}{2}\right) \int_{-\frac{\Gamma}{2}}^{\frac{\Gamma}{2}} d\varepsilon D(\varepsilon) f(\varepsilon + \Sigma_\sigma), \quad (9)$$

with $f(\varepsilon)$ being the Fermi distribution function. In order to focus on the spin ordering, we ignore the k -dependence of the self-energy. The summation with respect to \mathbf{q} is carried out exactly. The spin-ordered state is found by solving the following self-consistent equation:

$$\begin{aligned} m &\equiv \Sigma_\uparrow - \Sigma_\downarrow \\ &= C \int_{-\frac{\Gamma}{2}}^{\frac{\Gamma}{2}} d\varepsilon \sqrt{1 - \left(\frac{2\varepsilon}{\Gamma}\right)^2} \left[f\left(\varepsilon - \frac{1}{2}m\right) - f\left(\varepsilon + \frac{1}{2}m\right) \right], \end{aligned} \quad (10)$$

where $C = \sqrt{8/\pi} (e^2/\epsilon \ell_B) / \Gamma$.

At zero temperature, the condition for the quantum Hall ferromagnetic state is $\Gamma < \sqrt{8/\pi} (e^2)/(\epsilon \ell_B)$. This corresponds to the Stoner criterion for itinerant ferromagnetism in a metal. Using the parameter $\epsilon = 190 \text{ F/m}^{25}$ for α -(BEDT-TTF)₂I₃, we find $\Gamma < 5.4 \sqrt{B}$ with Γ measured in units of kelvin and B measured in units of tesla. When we consider that Γ is proportional to \sqrt{B} under high magnetic field, $\Gamma = \alpha \sqrt{B}$, where α is a constant. According to Tajima *et al.*,¹³ the Landau level width at $T = 1 \text{ K}$ is about 1.2 K. Below 1 K, the inter-layer magnetoresistance minimum exists at the magnetic field B that satisfies $2\mu_B B/\Gamma \simeq 1$. We define this B as B_0 . From the analysis of the experiment,¹³ we find that $\alpha \sqrt{B_0} \simeq 1.2 \text{ K}$ and $\alpha = 1.3$ at $T = 1 \text{ K}$. At lower temperatures, the parameter α appears to decrease. From this estimation of Γ , we may conclude that the Stoner criterion is satisfied. Meanwhile,

above 1 K, the width of Landau levels is mainly determined by the temperature and Γ is not proportional to \sqrt{B} . We surmise that α -(BEDT-TTF)₂I₃ is so clean that the \sqrt{B} dependence of Γ is not discernable. Therefore, here we take the elliptic density of state eq. (8) as a phenomenological formula, and take a constant value for Γ . If we consider the Zeeman energy and take $\Gamma = 2\mu_B B$, then we find that the Stoner criterion is satisfied for $B < 16$ T. Thus, we may neglect the effect of spin splitting for the intralayer spin-ordered states below $B = 16$ T. At finite temperature, we solve eq. (10) numerically. The result is shown in Fig. 1. From the interlayer magnetoresistance experiment,¹³ it was estimated that $\Gamma \sim 1$ K for $T < 1$ K. Therefore, we may conclude that the quantum Hall ferromagnetic state is stabilized within each layer in α -(BEDT-TTF)₂I₃ from Fig. 1.

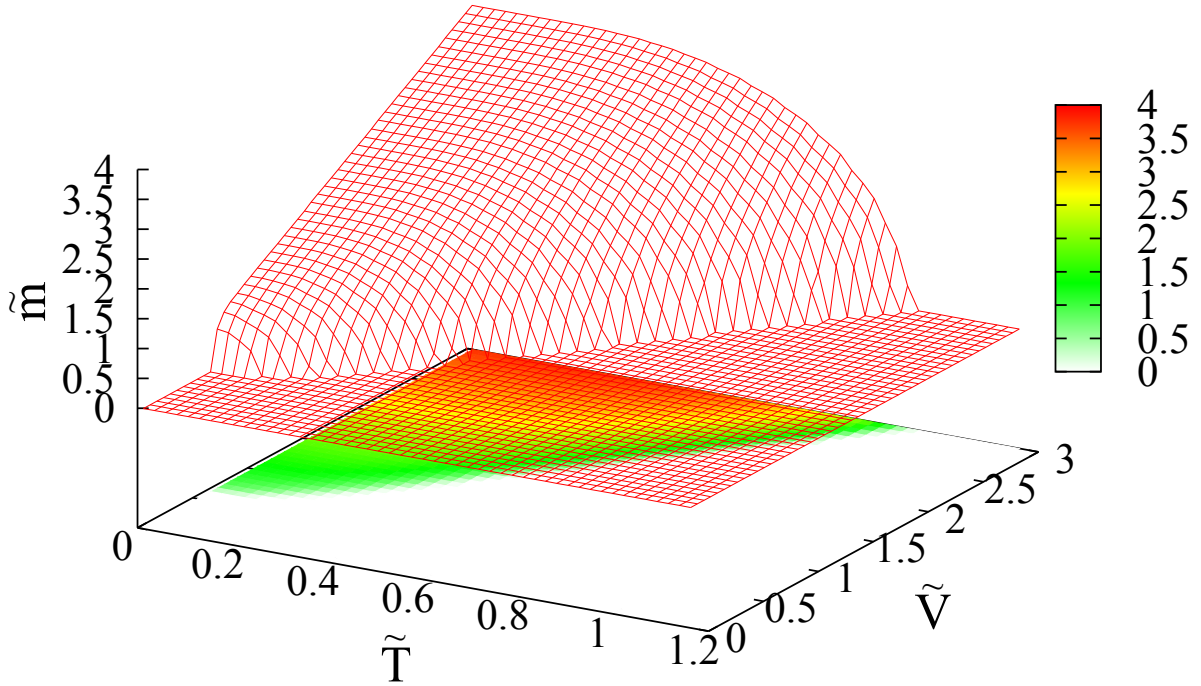


Fig. 1. (Color online) Temperature, $\tilde{T} = T/\Gamma$, and Coulomb interaction, $\tilde{V} = e^2/(\epsilon\ell_B\Gamma)$, dependence of the order parameter, $\tilde{m} = m/\Gamma$, for each layer of α -(BEDT-TTF)₂I₃ under pressure. We assume $\epsilon = 190$ F/m for the dielectric constant, which is inferred from the analysis of the interlayer magnetoresistance experiment.²⁵

The calculation above is easily extended to the $n \neq 0$ Landau levels, which is relevant for hole-doped α -(BEDT-TTF)₂I₃.²⁶ The difference is just the numerical factor of the Landau level form factors. However, there is not so much difference for the spin-ordering criterion itself between the $n = 0$ case and the $n \neq 0$ case. The critical Γ for the $n = 1$ Landau level is given by that for the $n = 0$ Landau level multiplied by the factor 11/16. However, we expect

that Γ takes large values in $n \neq 0$ Landau levels. Therefore, the quantum Hall ferromagnetic state may be unstable for high Landau levels.

Now we consider the interlayer coupling effect. When the condition for $m \neq 0$ is satisfied, each layer is in the spin-polarized state. Taking the z -axis for the direction of the spin polarization, we define $\hat{S}_{\mathbf{q}}^j \equiv (\hat{\mathbf{S}}_{\mathbf{q}}^j)_z$ by the Fourier transform of $\sum_{\sigma} \sigma \hat{\psi}_{j\sigma}^{\dagger}(x, y) \hat{\psi}_{j\sigma}(x, y)$, with j being the layer index. A crucial difference between graphene and α -(BEDT-TTF)₂I₃ is that the strong electron correlation plays an important role in α -(BEDT-TTF)₂I₃. In fact, the system is insulating owing to the strong electron correlation under ambient pressure.⁵⁻⁸ On-site Coulomb repulsion U and interlayer hopping t_{\perp} lead to the antiferromagnetic interaction, $J' = 4t_{\perp}^2/U$, between layers. The Hartree term associated with the interlayer antiferromagnetic interaction is

$$\begin{aligned} J' \sum_{j, \mathbf{q}} \hat{S}_{\mathbf{q}}^j \hat{S}_{-\mathbf{q}}^{j+1} &\simeq J' \sum_{j, \sigma, \sigma', \mathbf{q}, k, k'} \sigma \sigma' \exp(-q^2 \ell_B^2 / 2) \\ &\times \left(\langle c_{j, k, \sigma}^{\dagger} c_{j, k+q_y, \sigma} \rangle c_{j+1, k', \sigma'}^{\dagger} c_{j+1, k'+q_y, \sigma'} \right. \\ &\left. + c_{j, k, \sigma}^{\dagger} c_{j, k+q_y, \sigma} \langle c_{j+1, k', \sigma'}^{\dagger} c_{j+1, k'+q_y, \sigma'} \rangle \right). \end{aligned} \quad (11)$$

Hereafter, we only consider the zero energy Landau level, and we denote $\hat{c}_{0, k, j}$ as $\hat{c}_{k, j}$. We define the order parameter m_j for the j -th layer as $m_j = \sum_{\sigma} \sigma \langle c_{j, k, \sigma}^{\dagger} c_{j, k, \sigma} \rangle$, which is assumed to be k independent in accordance with the approximation introduced above. In terms of these order parameters, the Hartree term is rewritten as

$$J' \sum_{j, j', k, \sigma} \sigma \left(m_j c_{j', k, \sigma}^{\dagger} c_{j', k, \sigma} + m_{j'} c_{j, k, \sigma}^{\dagger} c_{j, k, \sigma} \right). \quad (12)$$

The interaction between spins in the j -th layer is

$$\begin{aligned} & - \sum_{\mathbf{q}, j, k, \sigma} V_{\mathbf{q}} \exp\left(-\frac{q^2 \ell_B^2}{2}\right) \langle c_{j, k+q_y, \sigma}^{\dagger} c_{j, k+q_y, \sigma} \rangle c_{j, k, \sigma}^{\dagger} c_{j, k, \sigma} \\ & \simeq -J \sum_{j, k, \sigma} (m_j \sigma + \rho_j) c_{j, k, \sigma}^{\dagger} c_{j, k, \sigma}, \end{aligned} \quad (13)$$

where ρ_j is the number density of the j -th layer and we defined $J = \sqrt{\pi/8} e^2 / (\epsilon \ell_B)$ for the intralayer ferromagnetic interaction parameter.

Including the Zeeman energy term the system is reduced to the following Ising model:

$$H_{MF} = \sum_j \sum_{i=1}^N \left\{ -J m_j s_j^i + J' (m_j s_{j+1}^i + m_{j+1} s_j^i) - \mu_B B s_j^i \right\}, \quad (14)$$

where s_j^i is the spin at the i -th site in the j -th layer. Note that the parameter J depends on the applied magnetic field B . Reflecting the fact that the interlayer coupling is antiferromagnetic,

the order parameter m_j takes different values for j even and for j odd. We denote the former and the latter as m and m' , respectively. The self-consistent equation for m, m' is

$$\begin{aligned} m &= \tanh [\beta (Jm - J'm' + \mu_B B)], \\ m' &= \tanh [\beta (Jm' - J'm + \mu_B B)]. \end{aligned} \quad (15)$$

Here, μ_B is the Bohr magneton and $\beta = 1/(k_B T)$ with k_B being the Boltzmann constant. We solve this self-consistent equation numerically and obtained Fig. 2 at $J' = 8$ K. Here, we assume a relatively large value for J' , which is the same order of magnitude as the interlayer hopping estimated in a related organic compound.²⁷ The parameter J' can be smaller depending on the ratio of the interlayer hopping to the on-site Coulomb repulsion. An antiferromagnetically spin-ordered state is possible only when $B = 0$ T. For $B > 0$ T, the spins are in a ferrimagnetically ordered state because of the Zeeman energy effect at low temperatures. The spin-polarized state is stabilized under high-magnetic fields where the Zeeman energy is larger than the interlayer antiferromagnetic interaction. The critical temperature T_c for the

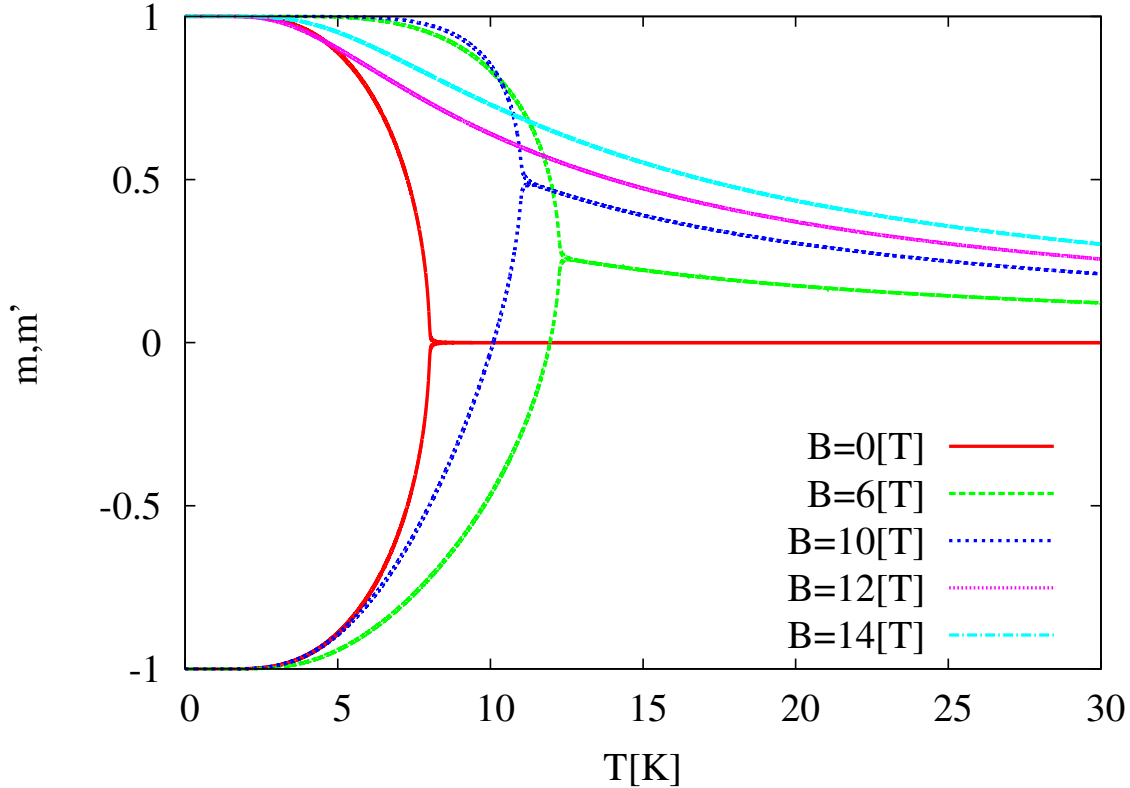


Fig. 2. (Color online) Temperature dependence of m and m' for different values of B at $J' = 8$ K. The ferrimagnetic states can be realized in the weak magnetic field regime. In the strong magnetic field regime the ferromagnetically spin-ordered state is stabilized.

magnetically ordering transition is obtained by substituting $m' = 0$ into eq. (15):

$$T_c = \frac{\mu_B B (J/J' + 1)}{\tanh^{-1}(\mu_B B/J')}. \quad (16)$$

Note that J depends on B . The phase diagram is presented in Fig. 3. The system is ferrimagnetic for $T < T_c$ and spin-polarized for $T > T_c$. When $J' < \mu_B B$, the antiferromagnetic interaction is irrelevant and only the spin-polarized state is stabilized. Even in the spin-polarized

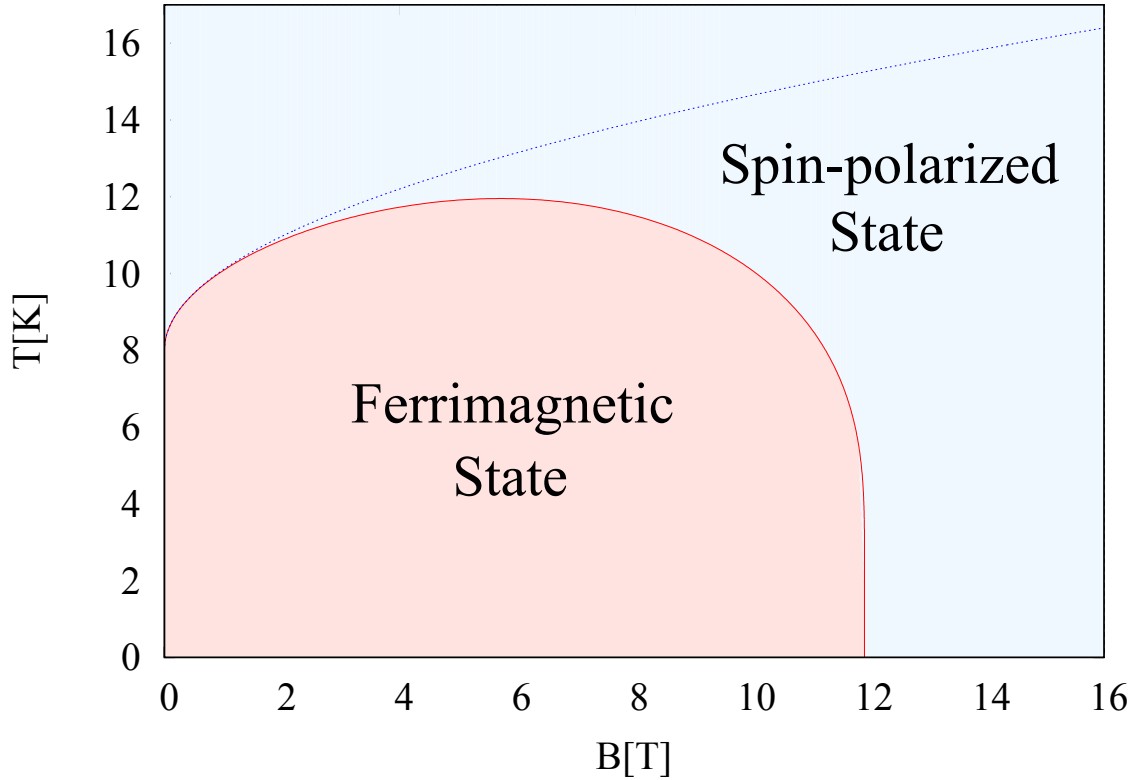


Fig. 3. (Color online) Phase diagram of spin-ordered states of the multilayer massless Dirac fermion system at $J' = 8$ K. The critical temperature for the interlayer ferrimagnetic state is given by the solid line, eq. (16). The dotted line shows the critical temperature for the interlayer antiferromagnetic state in the absence of the Zeeman energy.

state, a unique feature appears that is distinct from graphene. We introduce the effective g -factor as

$$g_{\text{eff}} = g + \frac{2}{\mu_B B} (Jm - J'm'), \quad (17)$$

with $g = 2$ being the g -factor in the vacuum. The temperature dependence of g_{eff} is shown in Fig. 4. Although the spins are ferromagnetically ordered, the mean fields associated with the neighboring layers suppress the energy splitting owing to the Zeeman energy because the

interlayer coupling is antiferromagnetic. As a consequence, g_{eff} is less than g . This behavior is in sharp contrast to that of graphene where the effective g -factor becomes larger than g .²⁸ This temperature dependence is consistent with the experiment in α -(BEDT-TTF)₂I₃.²⁹ To conclude, we have examined the spin-ordered states in multilayer massless Dirac fermion

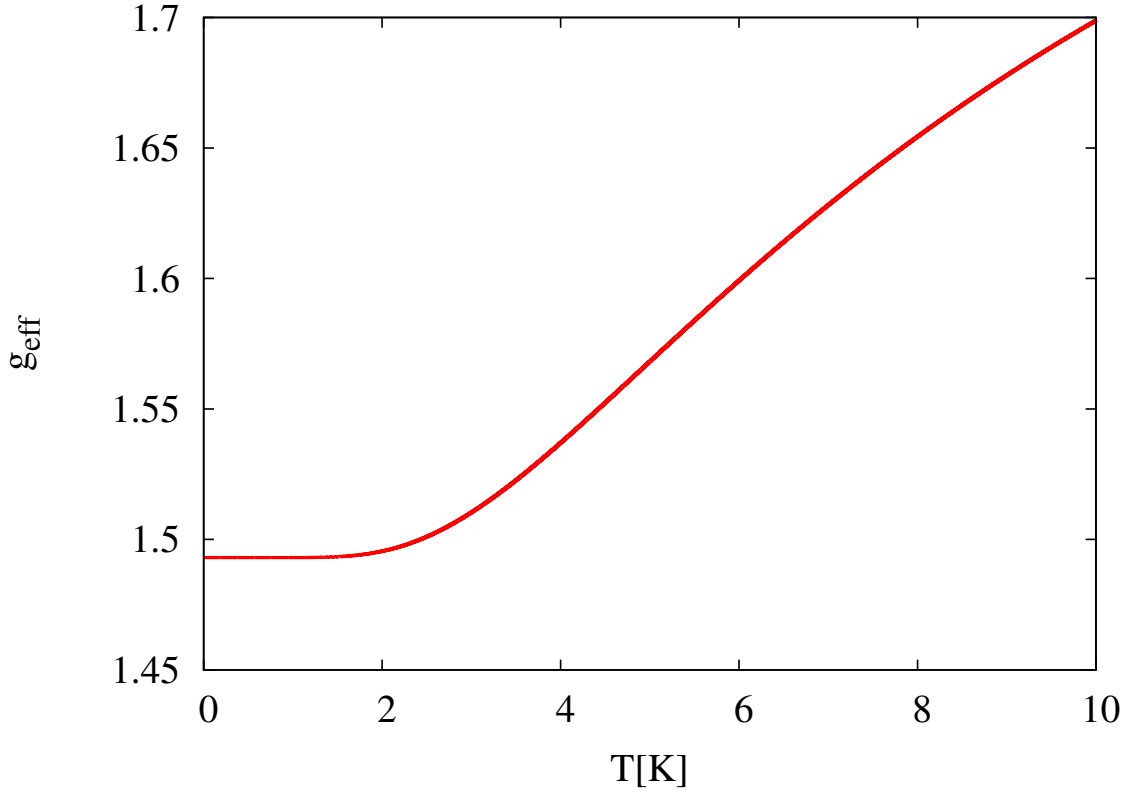


Fig. 4. (Color online) Temperature dependence of g_{eff} . The effective g -factor, g_{eff} , is less than $g = 2$ and decreases at low temperatures. This behavior is observed when $2.1 \sqrt{B} < J' < \mu_B B$ is satisfied.

systems. The exchange interaction leads to the ferromagnetic intralayer interaction while the strong electron correlation and the interlayer hopping lead to the antiferromagnetic interlayer interaction. Within the mean field theory, we have determined the phase diagram relevant for α -(BEDT-TTF)₂I₃. When the Fermi energy is at the Dirac point, the system exhibits the quantum Hall ferromagnetic state for $\Gamma < 5.4 \sqrt{B}$. The interlayer antiferromagnetic interaction leads to the ferrimagnetic state in the weak magnetic field regime. Even in a spin-polarized state, we expect an unusual behavior of the effective Zeeman g -factor, which is qualitatively consistent with the experiment.²⁹

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